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STUDY OF LOW-COST FABRICATION METHODS FOR AEROSPACE COMPOSITE MATERIALS

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January 1978

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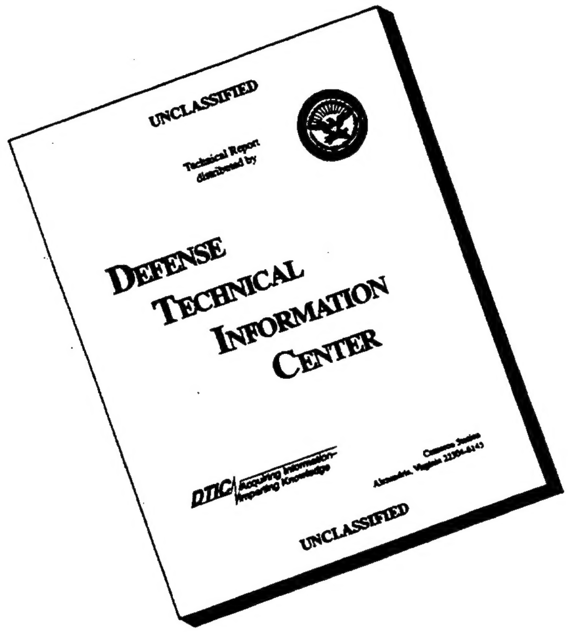

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STUDY OF LOW-COST FABRICATION METHODS FOR AEROSPACE COMPOSITE MATERIALS

FINAL REPORT

BY

HARVEY H. CHUNG

January 1978

PREPARED UNDER CONTRACT NASI-14730

BY

LOCKHEED-CALIFORNIA COMPANY
BURBANK, CALIFORNIA
A DIVISION OF LOCKHEED AIRCRAFT CORPORATION

FOR

NASA
NATIONAL AERONAUTICS AND
SPACE ADMINISTRATION

FOREWORD

This is the final report on study of low-cost fabrication methods for aerospace composite materials. It covers work completed between 6 January 1977 and 30 September 1977.

This program is being administered by the Langley Research Center, National Aeronautics and Space Administration with Mr. Edward L. Hoffman, group leader of the materials division as technical monitor.

The program was performed by the Lockheed-California Company with Harvey H. Chung the program leader, with assistance provided by R. F. Simenz and J. Wooley of the Materials and Producibility Department.

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ABSTRACT

Flat and hat section specimens of graphite/epoxy composite materials have been fabricated by the resin bath or wet pultrusion process and submitted to NASA-Langley. This demonstrated the feasibility of incorporating cross-plied graphite fiber reinforcements in conjunction with epoxy resin systems in the wet pultrusion process. However, the thickness constraints of the pultrusion process, the lack of dimensional stability of the crossply materials and equipment limitations affect the quality of the hat section pultrusions.

The wet pultrusion process shows promise of being a low cost method for producing composite parts with constant cross section along the length. A cost analysis showed at least 80 percent cost reduction for the hat section and 40 percent for flat panel by pultrusion over the conventional manual and automated lay-up.

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SECTION 1

INTRODUCTION

One of the most persistent factors which has limited the use of advanced composites in aircraft is the high cost for the weight saved. Pultrusion offers the potential of reducing the current cost of composite stiffener fabrication by at least 80 percent.

The pultrusion process combines the resins and reinforcements in a wet bath and then draws them through a heated die to convert them into finished products, continuously and automatically. Thus many of the intermediate labor and time consuming steps typically required with conventional composite fabrication methods are eliminated. However, in order to realize the full potential of this process, methods for introducing crossplies must be demonstrated.

The objective of this program was to demonstrate the feasibility of incorporating crossplied fiber reinforcements in the wet pultrusion process using existing pultrusion technology. The transverse reinforcement was to be provided using two methods: bias tape and mat. Flat panels and hat sections were fabricated using a $\pm 45^{\circ}$ bias graphite tape. Thickness constraints prevented the pultrusion of a 0.13 cm (0.052 inch) thick hat section, however, a 0.38 cm (0.15 inch) thick hat was produced with the crossplies. Attempts to use mat were not successful.

SECTION 2

RAW MATERIAL SELECTION

2.1 RESIN SYSTEMS

A study was made of available resin systems for wet pultrusion. The required system should have a minimum of eight hours pot life for regular production runs, low viscosity, fast cure at elevated temperature, good wetting, and mecahnical and physical properties comparable to today's structural composite resin systems. The only available epoxy resin that could be found was Aligned Fiber Corporation's AFC-902-B which was proven to be pultrudable and showed acceptable properties as shown on Table 1.

TABLE 1. GRAPHITE PULTRUSION TEST RESULTS

Graphite Fiber	Hercules AS
Matrix	AFC-902B Epoxy
Construction	$0^{\circ}/\underline{+}45^{\circ}$ = 55/45
Cross-Section Dimension	10.2 cm (4 in. wide) x 0.3 cm (0.12 in. thick)
% Resin Content	31.7
Specific Gravity	1.56
Flexural Strength	
at Room Temperature	1034 MPa (150 ksi)
at 180°F	655 MPa (95 ksi)
Flexural Modulus	
at Room Temperature	67.6 x 10^3 MPa (9.8 msi)
at 180°F	57.9 x 10^3 MPa (8.4 msi)
90° Flexural Strength	
at Room Temperature	208 MPa (30.3 ksi)
at 180°F	171 MPa (24.8 ksi)
90° Flexural Modulus	
at Room Temperature	14.5 x 10^3 MPa (2.1 msi)
at 180°F	13.1 x 10^3 MPa (1.9 msi)
Short Beam Shear	
at Room Temperature	77.9 MPa (11.3 ksi)
at 180°F	51.7 MPa (7.5 ksi)

2.2 GRAPHITE FIBER FORMS SELECTION

Graphite fibers used in the program were Union Carbide's Thornel 300 6K and Stackpole's Panex 30A which is a 7.62 cm (3 inch) wide unidirectional tape. The Panex tape had the heaviest tow available and required fewer spools to make up the needed fiber volume for hat sections. The selection of Thornel 300 fiber was based on the wide data base available at Lockheed.

2.2.1 Unidirectional Fiber Selection

Thornel 300 is available in 3K and 6K tows. The 6K tows have twice the amount of filament and are lower in cost. However, it would have taken 372 spools of 6K tows to make up the required amount of unidirectional fiber for the flat sections. At nominal weight of 0.454 kg (1 pound) per spool, this would have required 169 kg (372 pounds) and cost approximately $12,000 which far exceeded the allotted program funding. Furthermore, a huge creel or rack would have to be built to accommodate the numerous spools. With Union Carbide's assistance the required number of spools were reduced to 93 spools by collimating 4 tows of 6K onto one spool without additional cost. With this the purchase of graphite fiber and building of a creel was within the financial capability of this program. The Panex unidirectional graphite tapes were donated by AFC to make up the fiber volume needed for the larger cross-sectional area of the AFC hat section after many unsuccessful attempts to pultrude the thinner L-1011 hat section.

2.2.2 $\pm 45^{\circ}$ Graphite Crossply Material Development

The development of a crossply material suitable for pultrusion became a major task of this program. Extensive search and development work was devoted to obtain a crossply material having continuous length, dimensional uniformity and sufficient integrity to withstand the forces induced during the pultrusion. Such a crossply material was developed at Proform Inc. of Seguin, Texas and designated as DxG 075 graphite Knytex ($\pm 45^{\circ}$ graphite bias tape) and will be discussed in detail in the next section. The braided tape, an original candidate for crossply materials, was excluded from this program after considerable development effort in conjunction with the Woven Structures Company and Fabric Development Inc. to produce a 15.2 cm (6 inch) wide braided tape was

unsuccessful. A 10.2 cm (4 inch) wide tape is the maximum width which can be produced with the existing machine. However, Fabric Development Inc. did successfully produce a piece of 10.2 cm (4 inch) wide braided tape which has evenly distributed fiber as shown in Figure 1. In addition to the width limitation, the following reasons also inhibit its use in the pultrusion without extensive development:

1. Significant fiber degradation caused by the braiding process has been observed.
2. Uneven fiber tow distribution across the width of the tape as shown in Figure 1. The fiber tows are tighter on both edges and loose in the middle.
3. Significant tape thickness variation.

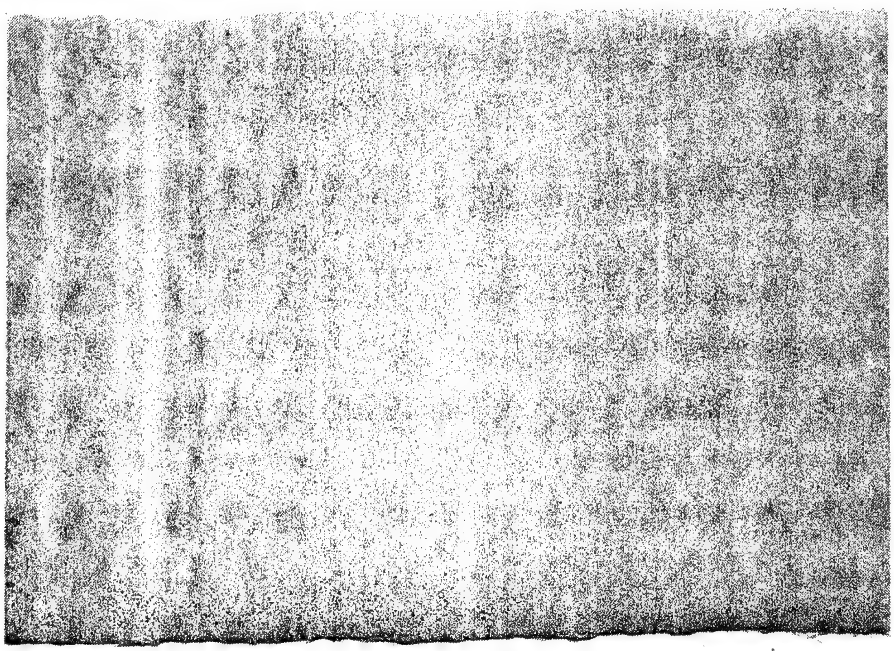

Figure 1. - Braided graphite tape supplied by Fabric Development Inc.

2.2.2.1 Development of $\pm 45^\circ$ Graphite Bias Tape

The bias tape was provided by Proform, Inc. in Seguin, Texas. It consisted of 18 picks of Thornel 300, 6K tows per 2.54 cm (1.0 inch), in the 45° direction supported by 6 stitches per 2.54 cm (1.0 inch) of a 40 denier polyester dacron yarns in the 0° direction (See Figure 2). Proform has designated this material Knytex.

The fiber orientation is easily distorted as a single ply, but combining two plies with a layer of epoxy prepregged lightweight scrim to get a $\pm 45^\circ$ tape provided the required dimensional stability. A fiberglass tape leader was attached to each roll of the bias graphite tape to start-up through the die. The combined $\pm 45^\circ$ bias tapes in the roll form are shown in Figure 3.

The Proform Kyntex material has several attributes for pultrusions and other composite structure as follows:

- The width of the bias tape can be made up to 356 cm (140 inches) wide.
- The desired number of dacron stitches can be programmed at any desired location and quantity.
- The weight of the tape can be determined by the number of picks per 2.54 cm (1 inch) of tows by selecting the proper machine gear ratio.
- Degradation of fibers during the stitching operation appears minimal compared to the braiding process.
- Thickness can be varied by tow size and number of picks per 2.54 cm (1 inch).
- The length of tape can be made up to 1829 meters (2000 yards) per roll
- The current cost of fabrication of the bias tape is about $22/Kg ($10/lb) over the graphite fiber cost.

2.2.2.2 Graphite Mat Development

Graphite mat was selected as the candidate for the lower cost crossply material for pultrusion since the fiberglass mat was widely and successfully used in the fiberglass pultrusion production. Consequently, graphite mat was believed to be a suitable choice for this pultrusion program. The only graphite mat immediately available at the beginning of this program was Union

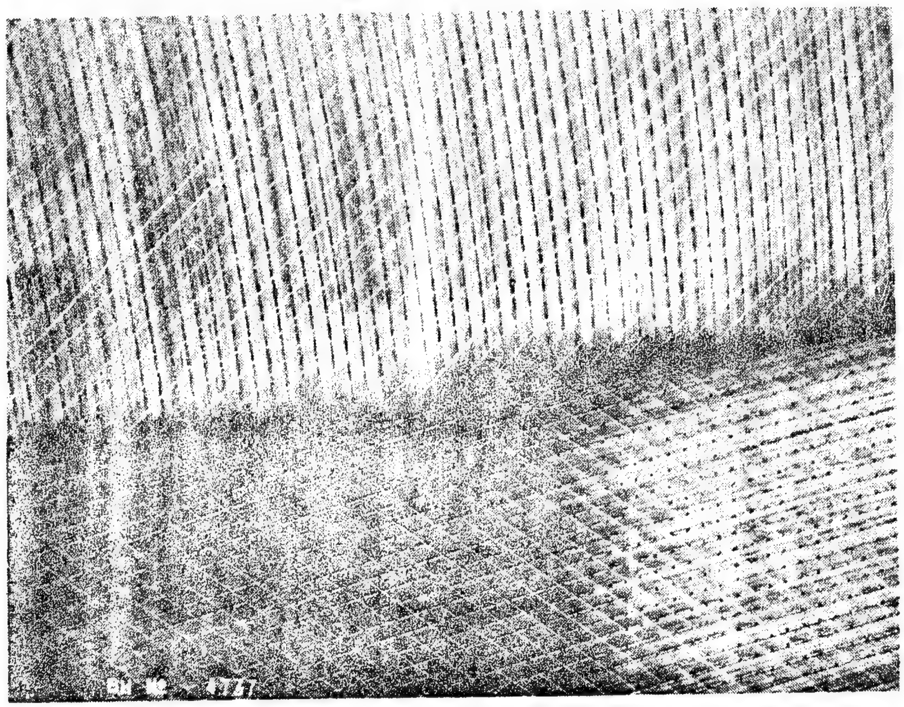

Figure 2. - 45° graphite bias tape supplied by Proform Inc.
Both sides shown.

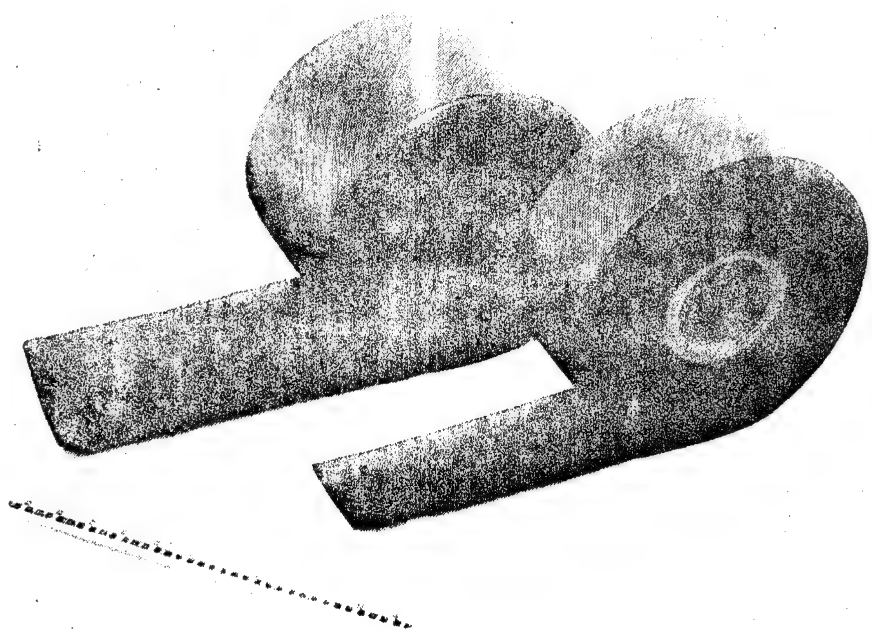

Figure 3. - ±45° graphite bias tape in roll form supplied
by Proform Inc.

Carbide's pitch fiber mat which is too bulky and loose to be pulled through a resin bath. Great Lakes Carbon Corporation developed a graphite web late in 1976 and claimed it would be available in early 1977 in development quantities. The sample of the web appeared to have good integrity and it was of proper thickness to facilitate good wetting in the pultrusion resin bath. However, the problems with precurser delivery, little enthusiastic response from other industries and heavy commitment of Great Lakes' machines for other graphite fibers have deterred its development.

Early this year Stackpole Fibers Company developed a series of graphite mats in various weights. The quality of the mat sample appeared to be suitable for pultrusion and it was decided to proceed with this material for the pultrusion program. Special mat material with a melamine binder had to be provided since the standard material utilizes a water soluble polyvinyl alcohol binder to hold the mat fibers together. The mat with melamine binder was delivered with a dacron scrim to give the graphite mat additional support.

SECTION 3

PULTRUSION DEVELOPMENT

The pultrusion work was conducted under subcontract to AFC, Inc., Chatfield, Minnesota. The graphite pultrusion effort for this program was performed on one of the production pultruders used for fabricating glass/polyester parts. An existing AFC pultrusion die was used to fabricate the 0.3 cm (0.12 inch) thick x 10.2 cm (4 inches) wide flat section and a hat section die designed to produce L-1011 composite vertical fin hat sections was fabricated to meet program requirements.

3.1 DESCRIPTION OF EQUIPMENT

The set-up for flat strip pultrusion consists of a creel, four resin tanks, guiding bars, die, pullers and cutter. The creel (or rack) holds 96 spools of Thornel 300-24K (4 ends of regular 6K tows) - 12 spools in a row and 4 rows on each side (see Figures 4 and 5). To produce the flat sections, three rolls of $\pm 45^{\circ}$ bias tape were held on a separate rack (see Figure 6) and each bias tape was combined with 24 tows of unidirection graphite fibers in its designated resin tank (See Figures 7 and 8). Four resin tanks were mounted vertically on a steel frame and a series of steel bars guide the graphite fibers to the die (see Figure 9). Figure 10 shows the final collimation of fibers before entering the die shown at the left side of picture and the resin puddle accumulated at the entrance of the die indicates good fiber wet-out. The die was clamped in between two steel plates which were heated by circulated hot oil.

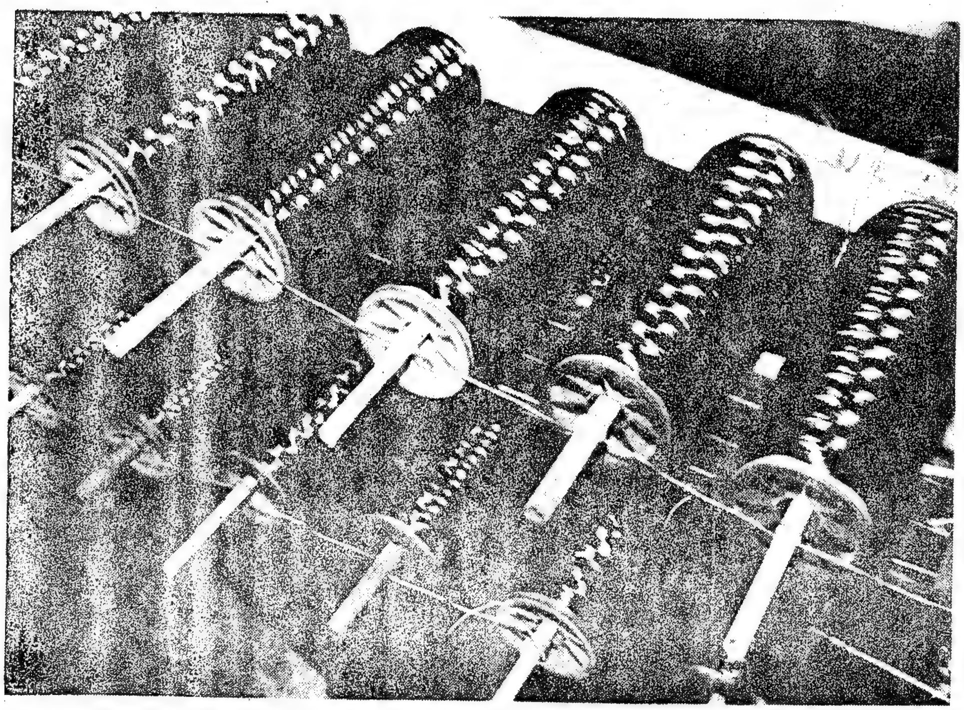

Figure 4. - Graphite fiber spools on creel

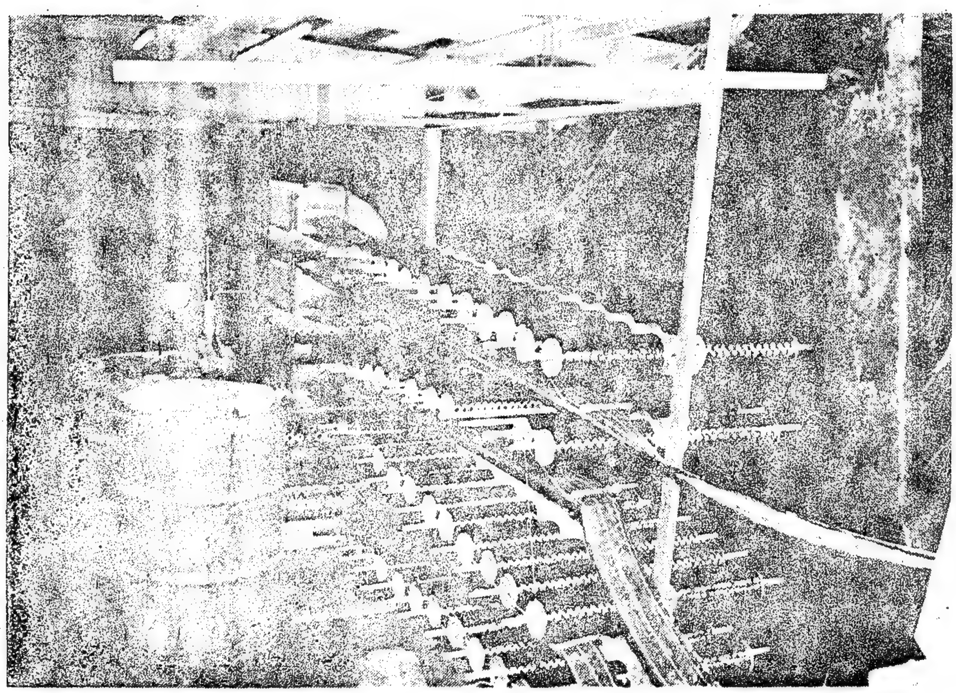

Figure 5. - Overall view of creel holding graphite fiber spools

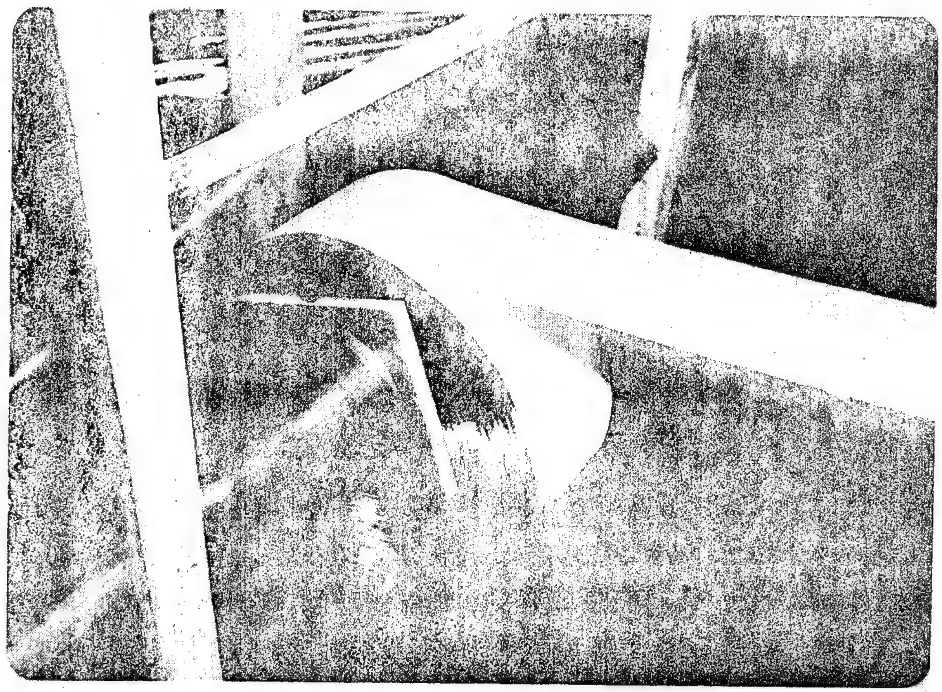

Figure 6. - $\pm 45^{\circ}$ bias graphite tape with fiberglass bias leader coming off the roll

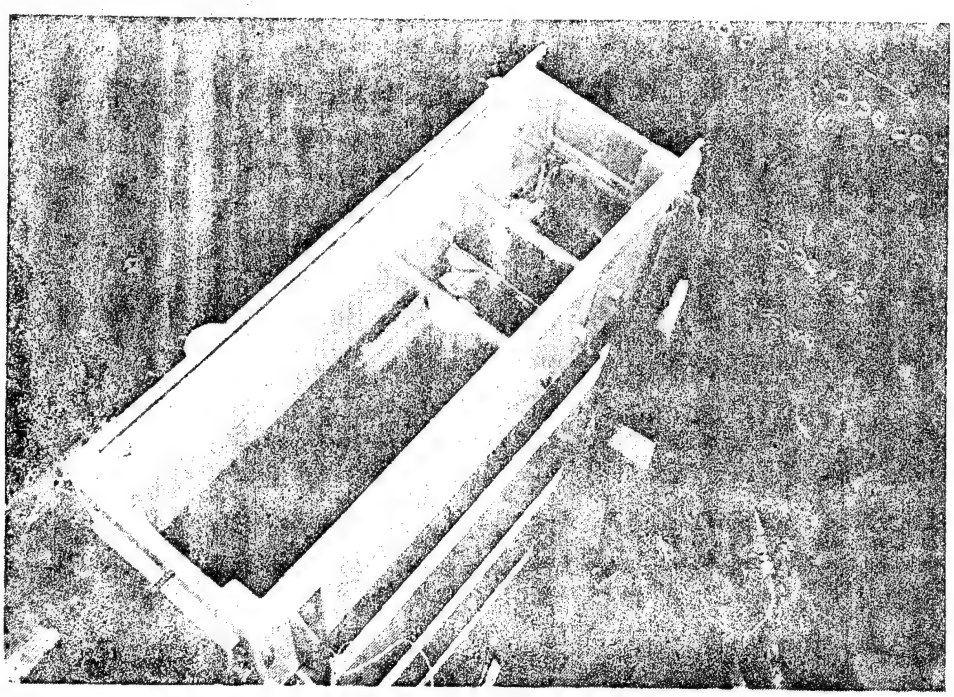

Figure 7. - Top view of the resin tank

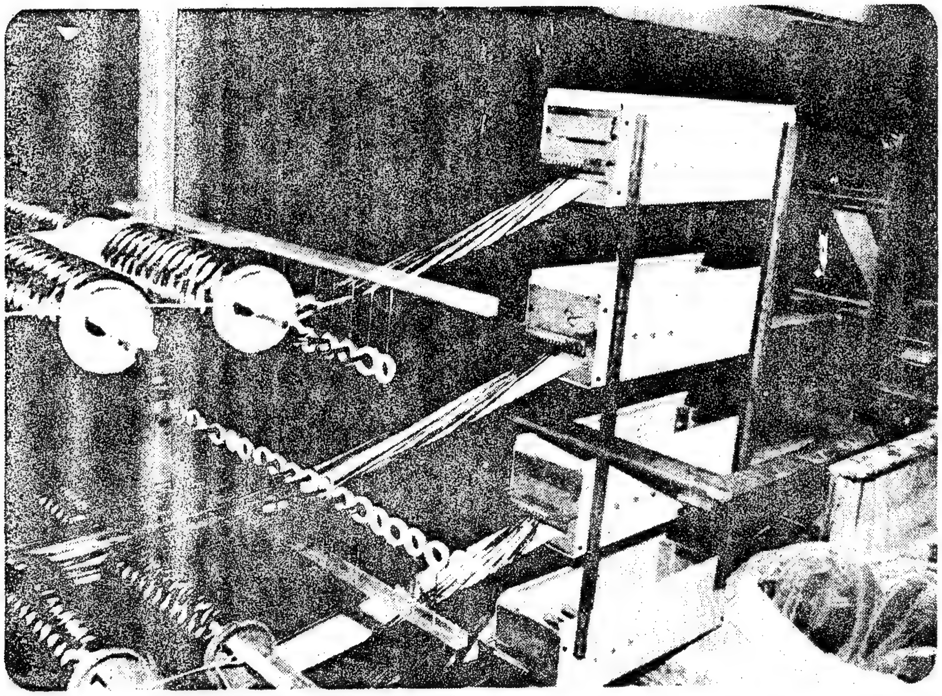

Figure 8. - Overall view of resin tanks

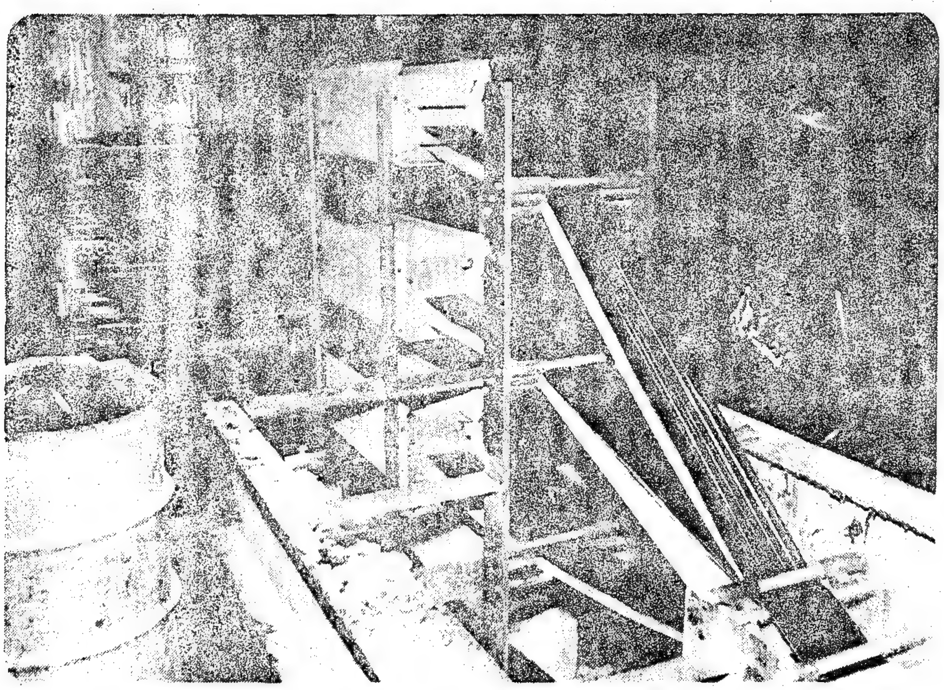

Figure 9. - Overall view of set-up before the pultrusion die

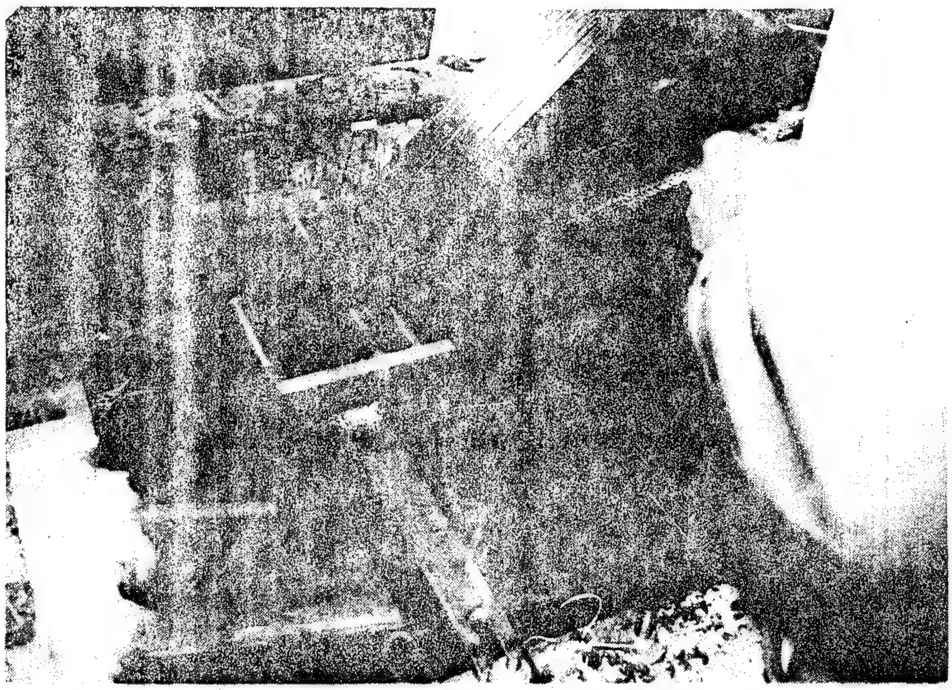

Figure 10. - The final collimation device before the pultrusion die

3.2 PULTRUSION WITH ± 45° BIAS TAPE

Trails run on three different ± 45° bias tape configuration were made before a successful flat section was produced. The first configuration consised of two layers of 45° Kyntex graphite tape sandwiched together with an epoxy resin preimpregnated glass scrim to form the ±45° bias tape. The unsuccessful since the polyester resin used to start up the operation apparently softened the epoxy on the scrim and allowed the ±45° tape to distort. The polyester resin was used in the initial stages of the pultrusion operation to facilitate the stopping and starting (at least 8 times) required to get the pultrusion through the die and to get the pullers operational. With epoxy resin, once the machine stops, it is very difficult to restart without breaking the pultruded strip. A second trial was made using ±45° bias tape which had been sewn together with three rows of stitches. However, the sewing operation had distorted the plies making the width uneven. Attempts to pultrude this material were also unsuccessful. Finally, a special epoxy was formulated for the scrim which would maintain its integrity when in contact with the polyester and

still be compatible with the epoxy pultrusion resin. Using 94 tows of Thornel 300-24K and two plies of the $\pm 45^{\circ}$ bias tape, 15.2 meters (50 ft) of the 10.2 cm (4 inch) wide x 0.3 cm (0.12 inch) thick flat section was successfully produced. The $\pm 45^{\circ}$ plies were sandwiched between the unidirectional fiber tows to facilitate the pultrusion operation.

Several attempts were made to pultrude an L-1011 composite vertical fin hat section using the $\pm 45^{\circ}$ bias tape. The hat section configuration is shown in Figure 11. It appears that with the existing equipment and pultrusion technology, uneven cross sections with thicknesses down to 0.13 cm (0.052 inch) cannot be pultruded due to bunch-up and breakage of the fibers. With more development, equipment modifications and minor design changes to allow for the pultrusion constraints, it is believed that hat sections similar to this configuration can be pultruded.

Based on the problems with the L-1011 hat section dies it was decided to use an existing AFC hat section die (see Figure 12) to demonstrate the feasibility of pultruding these shapes with crossplies. Forty feet of graphite hat section pultrusions were successfully pultruded with 96 tows of T300-24K graphite, 2 plies of $\pm 45^{\circ}$ bias graphite tape and 5 spools of 7.6 cm (3 inches) wide Panex 30A. The Panex was added since there was not sufficient spools of T-300 to provide the desired 55 percent fiber volume in the thicker 0.38 cm (0.150 inch) AFC die. The poor quality of the die affected the quality of the pultrusion in the cap area of the hat and the guidance system permitted distortion of some of the external unidirectional plies; however, these are equipment problems that can be resolved. Overall, the feasibility of pultruding thicker, constant thickness hat sections with cross plies was demonstrated. Factors affecting the relative success of the thicker hat section pultrusion are as follows:

- The constant thickness of AFC hat section provided constant pressure throughout the part.
- Thicker parts at 0.38 cm (0.15 inch) provided sufficient cushion to allow redistribution of the fibers in the die. This will have to be taken into consideration in the design of the part and the die.

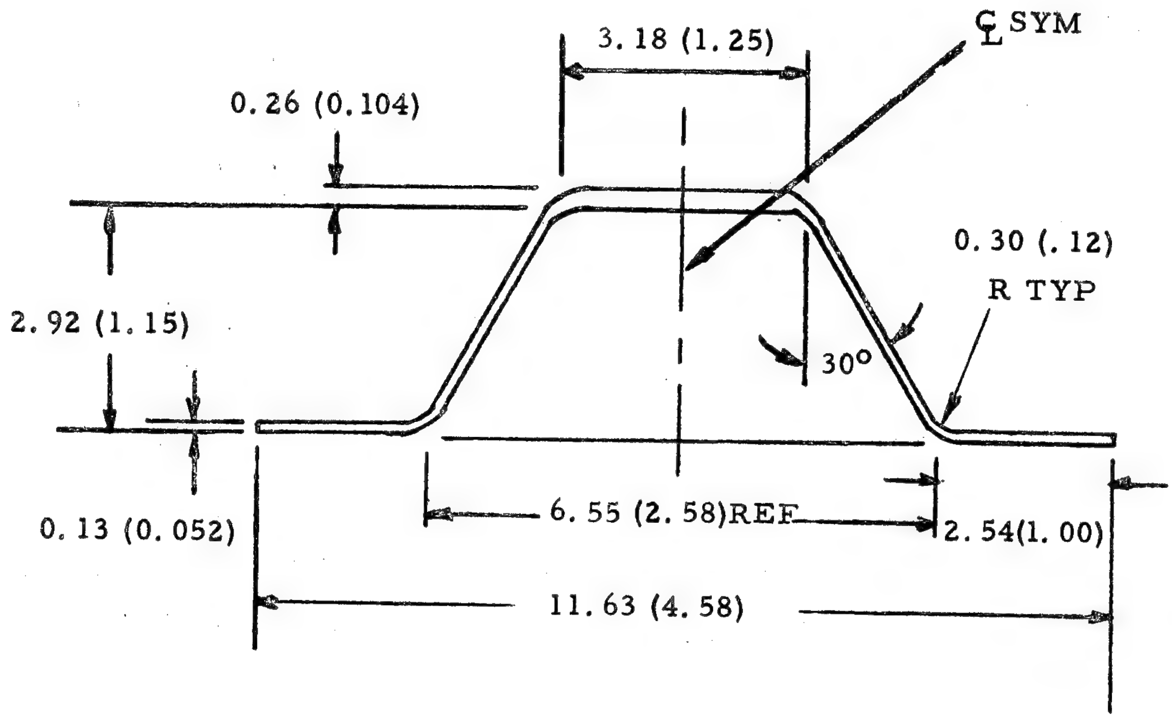

Figure 11. - L-1011 vertical fin hat stiffener, dimension in cm (in.)

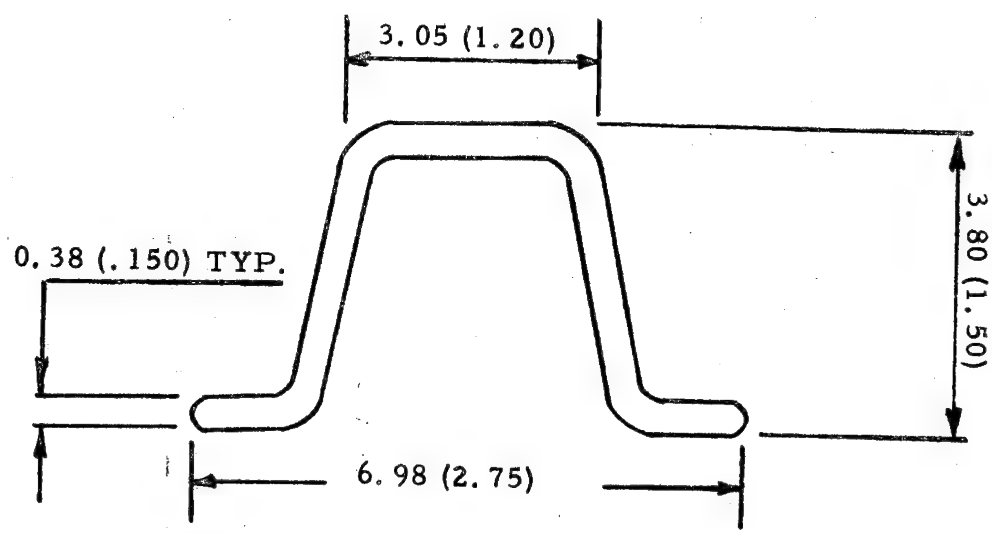

Figure 12. - AFC hat section, dimension in cm (in.)

- The die is only 61 cm (2 ft) long which is shorter than the L-1011 hat section die's 122 cm (4 ft) length. This allowed lower force for pulling the pultrusion through the two foot die.
- The bias graphite tapes were sandwiched between unidirectional graphite fibers.

3.3 PULTRUSION WITH GRAPHITE MAT

Another objective of the contract was to demonstrate the feasibility of providing transverse strength through the use of a more inexpensive material such as mat. The Stackpole's 0.33×10^{-2} g/cm^2 (1 oz/yd^2) and 1.0×10^{-2} g/cm^2 (3 oz/yd^2) graphite mats were evaluated at the end of a run by replacing the bias tapes with the mat. Only 4 plies of 1.0×10^{-2} (3 oz/yd^2) mat could be pulled through, and the line was subsequently broken by introducing two more plies of mat. The strips looked good, but the fiber volume was too low since 15 plies of the 0.33×10^{-2}g/cm^2 (1 oz/yd^2) mat are required to make 55 percent fiber volume strip. The problem is due to the bulkiness and lower integrity of the mat. The 0.33×10^{-2} g/cm^2 (1.0 oz/yd^2) mat was intended to improve the surface appearance of the pultruded strips and was discontinued due to poor appearance and poor integrity. New graphite mat must be developed which is thinner and stronger, if this approach is to be successful. The fiberglass mat commonly used in pultrusion industry does have these desired properties.

An attempt was also made to produce an L-1011 hat section using the fiberglass mat along with unidirectional graphite tows. The glass mats were placed on top and bottom directly against the die similar to the normal arrangement in the fiberglass/polyester pultrusions. As the mat entered the die it met tremendous resistance at the thin flange areas and bunched up at the crown area. The cured part barely made it through the die before the last section of the grippable dry fiber leader was broken. Despite many attempts to splice the broken leader, the spliced leader was no longer strong enough to overcome the tremendous friction between the solid part and the die as well as the resistance from jammed fibers. Disassembling of the die became inevitable. All further effort was discontinued due to the development requirements for pultruding with mat materials.

SECTION 4

COST ANALYSIS

The following analysis compares the labor and material costs incurred for pultruding versus the costs for automated laying up, two conventional cross-sectional shapes (a flat and a hat). The totals include fabrication labor and material costs only, stated in 1977 dollars. Quality Assurance sustaining labor, G and A, profit and other costs are not included. To assure a more meaningful analysis, both the conditions for fabrication in a developmental environment and a production environment are considered. Thus two sets of total costs are stated for each configuration and each manufacturing method employed.

ITEM I: FLAT - 10.2 cm (4 in.) wide x 0.3 cm (0.12 in.) thick

Premises:

Unit weight of flat section = 2.17 m/kg (3.24 ft/lb)
or = 459 g/m (140 g/ft)

- Percentage fiber volume = 55
- Percentage resin content = 30
- 0° graphite fiber needed = 150 g/m (45.7 g/ft)
- $\pm 45^{\circ}$ graphite bias tape needed = 150 g/m (45.7 g/ft)

Incremental Costs

- Bias tape = \$92.5/kg (\$42/lb)
- Thornel 300 6K graphite
  - (For developmental fab) = \$70.5/kg (\$32/lb)
  - (For production fab) = \$44/kg (\$20/lb)
- Resin
  - (For developmental fab) = \$260/15m (\$260/50 ft)
  - (For production fab) = \$4.4/kg (\$2/lb)
- Pultrusion
  - (For developmental fab) = \$740/15.2m (\$750/50 ft)
  - (For production fab) = \$13/m (\$4/ft)
- Graphite Prepreg
  - (For development fab) = \$114/kg (\$52/lb)
  - (For production quantity) = \$44/kg (\$20/lb)

Costs for Pultrustion:

	Development Cost		Production Cost	
	\$/m	\$/ft	\$/m	\$/ft
0° graphite tow	7.0	3.2	4.4	2.0
$\pm 45^{\circ}$ graphite bias tapes	13.8	4.2	13.8	4.2
Epoxy resin	2.1	0.6	0.7	0.2
Total raw material	22.9	8.0	18.9	6.4
Pultrusion cost (including labor and machine time)	48.6	14.8	13.0	4.0
Total	71.5	22.8	31.9	10.4

Costs for Flat Lay-Up (Using state-of-art fabrication technology)

	\$/m	\$/ft	\$/m	\$/ft
Material cost	41.00	12.50	16.41	5.00
Fab labor cost	105.00	32.00	42.44	13.24
Total lay-up cost	146.00	44.50	59.85	18.24

ITEM II: HAT SECTION

Premises:

Unit weight of hat section = 279 g/m (85 g/ft)
or = 3.6 m/kg (5.3 ft/1b)
Percentage resin content = 30
Percent fiber volume = 55
0° graphite fiber needed = 39 g/m (12 g/ft)
$\pm 45^{\circ}$ graphite bias tape needed = 154 g/m (47 g/ft)

Costs for Pultrusion:

	Development Cost		Production Cost	
	\$/m	\$/ft	\$/m	\$/ft
0° graphite tows	2.75	0.85	1.72	0.53
$\pm 45^{\circ}$ graphite bias tapes	14.25	4.35	14.25	4.35
Epoxy resin	3.20	0.40	0.50	0.15
Total raw material cost	20.20	5.60	16.47	5.03
Pultrusion cost (including labor and machine time)	48.60	14.80	13.00	4.00
Total	68.80	20.40	29.47	9.03

Costs for Hat Lay-Up (Using state-of-art fabrication technology)

	Development Cost		Production Cost	
	$/m	$/ft	$/m	$/ft
Material cost	47.57	14.50	18.87	5.75
Fab labor cost	533.16	162.50	164.44	50.12
Total lay-up cost	580.73	177.00	183.31	55.87

SECTION 5

CONCLUSIONS AND RECOMMENDATIONS

One notable difference between polyester and epoxy pultrusions is the surface appearance. A smooth and glossy surface is typical for the polyester pultruded parts in contrast to the rough and dull surface received with epoxy pultrusions. This is mainly due to the low adhesion and high shrinkage characteristics of the polyester resin that facilitate the easy release of the resin from the metal die surface during curing in the die. The epoxy resin tends to cling to the metal die surface due to its excellent adhesion to the metal. Furthermore the low shrinkage characteristic of the epoxy resin increases the difficulty of the resin release from the metal die. This is supported by the evidence of the cured epoxy residue remaining on the epoxy pultrusions. The difference in surface appearance may not make any difference in structural properties, nevertheless, it is an esthetic consideration.

The $\pm45^{\circ}$ bias graphite tape developed during this program appears to be an excellent crossply candidate for pultrusions. It is easy to handle and its fiber orientation remains relatively undisturbed in the cured pultruded parts. The correctly orientated fiber tows of the bias tape could be seen on the inside surface of the crown area of the hat section pultrusions. The bias tape exposed in this area is due to the unsophisticated guiding device which was not able to retain the intended 0° graphite fiber tow on the bottom of the cap area. The $\pm45^{\circ}$ bias tape provides the possibility for achieving high fiber volume composites pultrusion with little added development and it can be produced by a commercial machine at reasonable cost.

The graphite mats bulkiness and inability to hang together during pultrusion must be corrected before it can be used in pultrusions. More development is required to provide mat handling properties similar to those available with

fiberglass. The fiberglass mat was to be tried in this program after attempts with graphite mat were unsuccessful but funding constraints prohibited further effort.

Considerably more development is required before thin, uneven, cross sectional thickness similar to the L-1011 hat section can be pultruded using the wet process. However, the technology for producting flat sections and thicker shapes may be provided with only nominal development requirements.

An epoxy resin system possessing the needed pultrusion characteristics as well as the required structural performance properties for aerospace application must be developed. This is essential to the acceptance of pultrusions in aircraft design.

A cost analysis showed at least 80 percent cost reduction for the hat section and 40 percent for flat panel by pultrusion over the conventional manual and automated lay-up. These potential savings certainly warrant further development of the pultrusion process.

No physical or mechanical tests were conducted on the pultrusions produced for this program. NASA-Langely will perform this activity.